Thermal Ageing and Its Effects on the Stress Corrosion Cracking Susceptibility of Alloy182/A533B Dissimilar Weld Joints

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論文内容要旨

Stress corrosion cracking (SCC) incidents have been reported in Ni-base alloy weld portion in the A182/A533B dissimilar weld joint (DWJ) of core shroud supports in several BWRs worldwide. Concern has been raised on whether these cracks can grow into low-alloy reactor pressure vessel after longer operation time. On the other hand, it is well known that segregation of phosphorus (P) at prior-austenite grain boundaries (GBs) can cause temper embrittlement in low alloy steels exposed to temperature range 350 - 600°C. As a part of reactor pressure vessel in BWRs, core shroud support is a critical component of the pressure boundary. It is also a life-limiting component because the replacement is technically difficult and economically unviable. Although SCC susceptibility and P-induced temper embrittlement of low alloy steels have been extensively studied in the past years, little is known on the potential synergism between these two degradation modes. Since extending reactors service life up to 60 years or even 80 years is under consideration in many countries, it is necessary to understand whether synergism exists between thermal ageing and SCC in the DWJ, in order to assess the integrity of core shroud support during extended service life at BWR operating conditions. This research tackles this problem through three consecutive steps:

Firstly, segregation behavior in the heat-affected zone (HAZ) in both as-welded + post weld heat treated (PWHTed) and acceleratedly-aged DWJs were investigated using atom probe tomography (APT). Segregation level of principal segregants at grain/packet boundaries, precipitate-matrix interfaces and dislocations were characterized qualitatively and quantitatively. It was found that non-equilibrium segregation dominated welding/PWHT and subsequent cooling process, leading to microstructure-dependent segregation of solutes at boundaries. Equilibrium segregation occurred during accelerated ageing, resulting in further increase of P and Mn segregation, but desegregation of C and Mo at boundaries.

Secondly, SCC behavior in the HAZ of both as welded + PWHTed and acceleratedly-aged DWJs were investigated using creviced bent beam tests in oxygenated high-temperature water doped with SO_4^{2-} and Cl^-. Crack retardation, propagation and

reactivation in HAZ were characterized in detail. Correlation between P segregation and SCC was established, suggesting intergranular and transgranular SCC have probably been promoted by P segregation at grain/packet boundaries and precipitate-matrix interfaces, respectively.

Finally, evolution of P segregation at GBs over long-term ageing at LWR operating temperatures is modeled by McLean's model based on findings in APT study. The results were correlated to susceptibility of temper embrittlement by empirical formulas, suggesting significant increase in the ductile-to-brittle transition temperature might only be expected in very limited region in HAZ. The results of this research suggest that thermal ageing could reduce SCC resistance in LAS, but only to a limited degree that is unlikely to cause serious concern to the overall integrity of the core shroud support after extended service life.

Fig. 1 outlines the completed research.

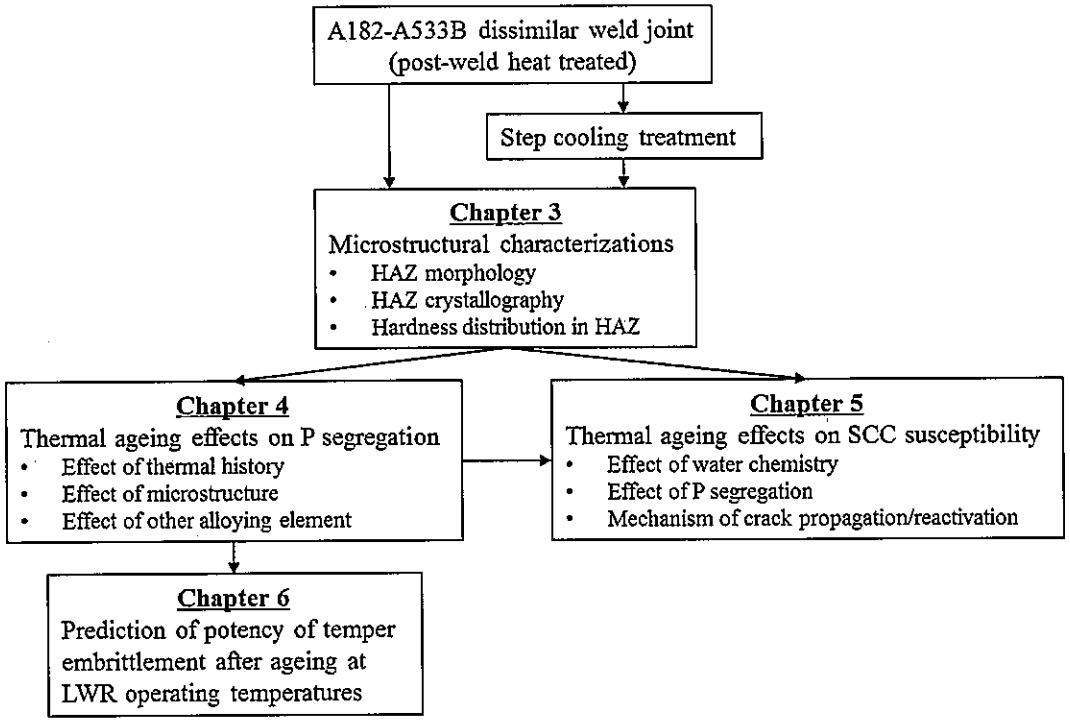

Fig. 1: Outline of the research performed.

In total, the thesis consists of seven chapters. The main content of each chapter is described below.

Chapter 1 introduces the current status and challenges of the life extension of light water reactors, from which this research was necessitated. Bibliographic review on related issues is also present, with objectives and approach being specified.

Chapter 2 describes the materials, experimental techniques and analysis methods that are used in this research. In particular, specimen preparation and experimental procedures are detailed.

Chapter 3 summarizes the microstructural characterizations of the DWJ necessary to understand the microstructural inhomogeneity resulted by multi-pass welding. Chemical etching revealed that complex transformation had taken place in the

immediate vicinity of fusion boundary, which can be classified into coarse-grained HAZ (CGHAZ), fine-grained HAZ (FGHAZ) and intercritically reheated CGHAZ (ICCGHAZ). Crystallographic features and hardness distribution in these three types of HAZ microstructure are displayed.

Chapter 4 presents the thermal ageing effects on P segregation in HAZ studied using atomic force microscopy (AFM) and APT. APT is mainly used to characterize the segregation behavior in real HAZ both with and without accelerated ageing, with focus on the effects of thermal history and microstructure on the segregation of P, as well as of other alloying elements that may interact with P during segregation. Results of qualitative and quantitative analysis are presented. Dominant mechanism of segregation during fabrication and accelerated ageing process, as well as the influencing factors of P segregation at grain/packet boundaries are clarified, with corresponding reasoning being described in detail. In addition, segregation of solutes at locations other than GBs is also characterized, and its potential consequences on P segregation during ageing are discussed. On the other hand, since APT can only analyze segregation behavior at very limited region of an interface, AFM was also employed to examine P segregation at a more general scale. The depth of GBs etched by immersing in picric acid-based solution was measured as indicator of segregation level of P at GBs, and was correlated to the data measured using APT to check the representativeness of APT data. The result of this process is also presented in this chapter.

Thermal ageing effects on SCC susceptibility of HAZ at simulated BWR conditions investigated by CBB tests are presented in Chapter 5. Morphology of SCC in A182 and in HAZ are shown. Qualitative and quantitative results of the SCC behavior in base material and DJW both with and without ageing are presented and compared. Models for SCC propagation/reactivation in the immediate vicinity of fusion boundary in HAZ are proposed, along with relevant mechanisms of high likelihood. Extrapolation between P segregation and SCC susceptibility is given, enabling quantitative estimation of the effects of P segregation on intergranular SCC in HAZ.

Chapter 6 details the modelling to predict evolution of P segregation in HAZ over long-term ageing at LWRs operating temperatures. Ideal equilibrium segregation in Fe-P binary system without site-to-site interactions is presented. Selection of values of the coefficients necessary for calculation is described. Results are presented, and their implications on the integrity of core shroud support over long-term ageing are discussed.

Chapter 7 summarizes the experimental findings and key conclusions of this research. High-priority items for future work are also suggested.

論文審査結果の要旨

世界的に発電用軽水炉の高経年化が進む中で、今後顕在化する可能性のある経年的材料劣化を予見的に評価しておくことが、軽水炉の長期信頼性を保証する上で極めて重要である。本論文は、軽水炉の1次系圧力バウンダリの中でも最も重要な機器である原子炉圧力容器について、炉水温度での熱時効が低合金鋼の溶接熱影響部の不純物偏析に及ぼす影響を明らかにし、その材料特性への影響を論じたものである。

第1章は序論であり、本研究の背景と目的、意義を述べている。

第2章では、本研究で用いた材料、試験・測定機器、試験方法について、原理的説明から独自に開発した試験手順に至るまで詳しく述べている。

第3章では、沸騰水型軽水炉圧力容器の炉底部に存在する 182 合金／A533B 鋼異材溶接継手を模擬した試験体の微視組織について、とくに本研究で注目している低合金鋼側溶接熱影響部のキャラクタリゼーション結果を述べている。溶接時の多重熱サイクルにより形成される熱影響部組織を金属組織学的特徴に基づいて分類し、それらの分布を明らかにしている。

第4章では、化学的エッチングと3次元アトムプローブを併用して、熱時効によるリンの粒界偏析挙動を明らかにしている。原子炉圧力容器用鋼溶接熱影響部のリン偏析感受性は微視組織に強く依存し、インタークリティカル組織、粗粒組織、細流組織の順にリン偏析傾向が高いことを明らかにした。また、粒界へのリン偏析は、施工時の溶接熱あるいは溶接後熱処理による非平衡偏析ならびに低温長時間時効における平衡偏析の双方があり、粒界の濃度プロファイルの特徴が異なることを、アトムプローブを用いた実測により明示した。さらに、炭素、マンガン、モリブデンとリンとの共偏析あるいは競合偏析の関係を、非平衡偏析および平衡偏析の場合についてそれぞれ明らかにした。これらは、本研究により初めて明らかにされた重要な知見である。

第5章では、熱時効が高温高圧水中での応力腐食割れ感受性に及ぼす影響を実験により評価し、182 合金側から入り 182 合金／A533B 境界で一旦停留したき裂が、再び進展を開始する傾向の大小に着目して、時効による応力腐食割れ加速効果とリン偏析関与の可能性を初めて指摘した。

第6章では、第4章で実測されたリン偏析挙動と広範な文献データに基づいて、炉水温度で長時間時効を受けた場合の原子炉圧力容器の脆化を予測している。溶接熱影響部のインタークリティカルおよび粗粒組織においてのみ際立って脆化感受性が高く、とくに加圧水型炉では延性－脆性遷移温度の上昇が無視できない可能性があることを初めて指摘している。

第7章は結論であり、本論文の成果を総括するとともに、今後取り組むべき研究の方向を整理して述べている。

以上、本論文は、原子炉圧力容器の長期信頼性確保の枢要である溶接熱影響部を対象として、3次元アトムプローブ等による実測と解析的予測を用いて低温長時間時効による粒界等への不純物偏析現象を初めて詳細に明らかにし、あわせてその高温水中応力腐食割れ感受性への影響を初めて指摘したものである。高経年化が進行する軽水炉の経年的材料劣化に対して的確な予測的対応をとる上で極めて重要な知見を多数与えており、量子エネルギー工学の発展に寄与するところが少なくない。

よって、本論文は博士(工学)の学位論文として合格と認める。